ISOTOPES AS INDICATORS OF RADIONUCLIDE MIGRATION AWAY FROM UNDERGROUND NUCLEAR TEST CENTERS

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Between 1955 and 1992 the United States conducted 825 underground nuclear tests at the Nevada Test Site (NTS) in southern Nevada. Higher yield tests were fired in Tertiary silicic volcanic rocks of Pahute Mesa; lower yield events were fielded in Quaternary alluvium and Tertiary volcanics of Yucca Flat. Approximately one-third of all the experiments were detonated below or within one cavity radius of the static water level. The potential for groundwater contamination associated with nuclear testing has long been recognized. In this context, a distinction must be drawn between the radionuclide source term which includes all radioactive material remaining after a nuclear test and the hydrologic source term which includes only those radionuclides dissolved in and transported by groundwater. Risk assessment developed for human health and the environment requires a reliable measure of both radionuclides available for potential transport by groundwater and probable groundwater flow-paths to down gradient receptors.

Objectives of a radionuclide migration program are threefold: 1) accurately measure the radionuclide source term for all underground tests at the Nevada Test Site, 2) determine the release potential of radionuclides to groundwater and the evolution of the hydrologic source term and 3) use isotope measurements to develop realistic conceptual and numerical models of groundwater flow and radionuclide transport.

Together with the Los Alamos National Laboratory, the Nuclear Chemistry Division of Lawrence Livermore National Laboratory has completed an inventory of fifty-four long-lived radionuclides residual from underground nuclear testing including residual fissile fuel and tracer materials, fission products of fuel burn, activation products induced by neutron reactions with device parts and the surrounding geologic medium and tritium. Event-by-event radionuclide totals are published in a classified format; regional totals corresponding to five NTS test areas are proposed for unclassified publication (Goishi et al., 1994). Radionuclide totals are decay corrected to January 1, 1994 and January 1, 2094. A compilation of specific radionuclide totals for the entire Nevada Test Site is decay corrected to January 1, 1994 and tabulated below. This list represents selected fission products, actinides and tritium important to the inventory together with a measure of the natural radioactivity introduced by melting of evolved volcanic rocks.

Total NTS Radionuclide Inventory in Curies
Decay Corrected to January 1, 1994

^{3}H	^{85}Kr	^{99}Tc	^{137}Cs	^{235}U	^{239}Pu	Nat'l U
1.17×10^{8}	1.64×10^{5}	5.70×10^{2}	2.77×10^{6}	2.40×10^{2}	7.53×10^{9}	5.95×10^{1}

The release of radionuclides to groundwater produces a hydrologic source term and bounds contaminant fate and transport. Summaries of experiments involving radionuclides leached from nuclear explosive melt glasses under saturated and partially saturated conditions indicate decreasing leaching rate averages (in fraction leached per day): I > Te > W > Sb > Cs > U > Co > Ru > Ta > Ba > Ce > Mn Y > Zr > Pu (Smith, 1993).

End-member compositions enriched in alkaline earths and $SiO_2 + Al_2O_3$ were determined with the electron microprobe. Leaching rate is dependent on the major element composition of the glass; alkali and alkaline earth glasses are more susceptible to dissolution than those enriched in silica and alumina. The nuclear explosive debris consists of a heterogeneous mixture of quartz, plagioclase, potassium feldspars and glass. Refractory radionuclides (Am, Cm, Pu, Ce, Eu and Zr) are concentrated in the glass; volatile species are associated
with both glass and crystalline phases. Sorption coefficients for NTS tuffs and alluvium may be arranged in decreasing order: Eu > Ce > Y > Co > Cs > Ba > Nb > Ru > I > U > Sr > Sb; in general Pu > Cs, Cs > Sr and Sr > Tc (Smith, 1993).

Stable, cosmogenic and radiogenic isotopes have been measured in concert on NTS groundwaters to conceptually model sub-surface flow (Davisson et al., 1994). The hydrostratigraphy of the NTS consists of a regional Paleozoic carbonate aquifer and aquitard unconformably overlain by Tertiary tuffs and rhyolite lava aquitards and aquifers. The stratigraphic section is capped by Quaternary basin fill alluvial aquifers. Depth to the static water level at NTS ranges between 100 and 625 meters. Groundwater measured from NTS wells have $\delta^{18}O$ values that range from -15.0 to -12.5 ‰ with the most depleted values found in the higher mesas and heavier values measured in the lower valleys. In general there is enrichment in $\delta^{18}O$ values in groundwater from recharge areas to the northeast to discharge areas in the southwest; this is consistent with flow directions inferred from water table maps. The majority of recharge to the NTS is apparently derived from regional flow. ^{14}C abundances in NTS groundwaters range between < 1 to 60 percent modern carbon.

Young apparent ^{14}C groundwater ages (modern to 20,000 years) are typically observed under Pahute Mesa; older apparent groundwater ages (4100 to > 40,000 years) are observed beneath Yucca Flat. The low groundwater flow rate into Yucca Flat is controlled by low permeability aquitards that surround its upgradient side and is reflected in an abnormally deep water table. Groundwater flow under Pahute Mesa has less lithological restriction and is controlled mostly by fracture permeability in the volcanic section. Measured $^{36}Cl/Cl$ ratios in NTS groundwater range from 100 to 1000 x 10^{-15} and their variation is controlled by introduction of "dead" Cl by water-rock interaction along vadose and saturated zone flow paths. Analyses of $^{87}Sr/^{86}Sr$ ratios of groundwater sampled from the Paleozoic carbonate rocks and Precambrain siliciclastic rocks indicate that radiogenic signatures as high as 0.7151 result from water-rock reaction. These higher ratios contrast with groundwaters sampled from Tertiary volcanic rocks that range between 0.7055 and 0.7095.

The rates and mechanisms of radionuclide migration from test cavities are linked to local and regional groundwater sources and flow quantified with the combined isotope approach. The research results will provide conceptual input into a system model developed for groundwater risk assessment and monitoring over the southern Nevada region.

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